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Time-Resolved Laser-Induced Incandescence Measurements for the EPA Heavy-Duty Federal Test Procedure

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ABSTRACT

Laser-induced incandescence (LII) is a promising new diagnostic for measuring the volume fraction of elemental carbon in engine exhaust. The technique is considerably more precise and sensitive than conventional measurement procedures, and can be applied either with or without dilution. However, LII has been slow to gain acceptance because of presumed complexity of use and high initial cost. In this paper we demonstrate a prototype LII system that offers turn-key operation and long-term cost that is highly competitive with other techniques because of very low labor costs. The LII system ran unattended for 7.5 weeks, logging 1078 heavy-duty diesel engine tests during 24/7 operation of a dilution tunnel facility. Among the tests logged were 363 FTP steady-state mode tests and 250 FTP transient tests for which gravimetric measurements of total particulate matter (PM) were obtained. Of these tests, removal of the filter-based volatile matter using supercritical fluid extraction was performed on 142 and 147 of the tests, respectively. The correlation between the time-integrated LII signals and the dry gravimetric measurements for the steady-state mode tests is used to calibrate the LII measurements in mass units. This calibration is then used to evaluate the correlation between the LII and dry gravimetric measurements for the transient tests. Finally, time-resolved LII measurements for the steady-state mode tests are presented to illustrate three forms of unsteadiness that would seem undesirable.

INTRODUCTION

Laser-induced incandescence (LII) has been shown over the past few years to be a valuable measurement technique for characterizing diesel particulate matter (PM) emissions. A high-energy, pulsed laser is used to heat the PM to very high temperatures, resulting in blackbody radiation that is proportional to the volume concentration of elemental carbon (EC). Key advantages of LII over more established, conventional techniques, are the ability to make real-time measurements in raw exhaust (i.e., without dilution) with high sensitivity (limit-of-detection to better than one part-per-trillion by volume [1] or 20 $\mu g/m^3$ [2]). In addition, because an LII system is virtually maintenance free, it is possible to run continuously, producing an analog-out signal suitable for a conventional, emissions data-acquisition system.

Three research groups have been particularly active in the development of LII for reciprocating engine exhaust applications, located at the Friedrich-Alexander-Universität, Erlangen-Nürnberg [3-5], the National Research Council (NRC) Canada [6-8], and the Sandia National Laboratories [9,10]. The group in Germany is affiliated with a private company, Esytec Gmbh, which has offered a commercial LII instrument for several years [11]. The groups at NRC and Sandia also have commercialization as a goal, and are partnering with a private company, Artium Technologies, Inc., to produce a commercial instrument [12]. Finally, a group at Argonne National Laboratory is also working toward commercialization of an LII instrument for exhaust measurements [13]. The fact that no commercial sales of an LII instrument have occurred as of the start of 2004 is testimony that the cost-to-benefit ratio is deemed excessive. However, we believe that assessment by initial capital cost is shortsighted since, if labor costs are included in the evaluation process, LII becomes very competitive. We estimate that the capital cost of an LII instrument would be recovered in approximately 3-6 months of unattended 24/7 operation

LII SYSTEM

The measurements presented were obtained at the Cummins Technical Center using Sandia's portable LII system, which is essentially a development laboratory packed onto a 2′x4′ cart. It contains the Nd:YAG laser and power supply for LII excitation, signal detector, mini-tower PC, optical cell for access to the exhaust flow, and blower to extract the exhaust sample from the constant volume sampling (CVS) dilution tunnel. The only external connections to the cart are 110 volt/15 amp power, 30′ heated exhaust-sample line, and vent line.

The normal mode of operation of the LII system is to record individual tests on the hard drive of the cart PC. This procedure requires a dedicated operator to create data files and start and stop each test, and additional post-test processing to integrate the cart data with the measurements acquired by the native Cummins data-acquisition system. To simplify this process, a digital-to-analog conversion of the LII measurements was sent to the native system, where it was logged onto an available analog-to-digital channel. The LII system was configured to run continuously in a totally passive mode, such that whenever a test was logged by the native system the LII signal was available. The LII system ran in this mode for 7½ continuous weeks, except for one inadvertent disconnection of power that was not discovered for several days. The major issues regarding durability of the LII system are flashlamp lifetime and fouling of the optical-cell windows. We estimate that the flashlamp fired 60 million times during the test period, with no measurable deterioration in laser energy (a replacement lamp costs less than $200). An active air-purge kept the cell windows spotless during the entire test period.

The laser energy is set during a “typical” steady-state test to the level that gives the maximum peak in the instantaneous LII signal. At this condition the EC has reached the sublimation temperature without measurable volume loss having yet occurred. Nanosecond-resolution data acquisition is required to capture this maximum of the temporal LII signal, which is directly proportional to the EC volume concentration. The digital-storage oscilloscope used for data acquisition has a 500 MHz bandwidth with a 5 GigaSample/s digitization rate. Communication between the PC and the oscilloscope is done using GPIB (general purpose interface bus) commands, which are very slow by contemporary PC standards. In addition, there are periodic “overhead” operations performed by the oscilloscope that cannot be defeated. As a result, even though our laser runs at a 20 Hz repetition rate, the actual data acquisition rate was limited to approximately 3 Hz.

TEST FACILITIES AND METHODOLOGY

The tests were carried out at Cummins Inc.’s Transient Emissions Test Facility. Engine exhaust from either of two test cells was directed to an 18 inch diameter full flow Constant Volume System (CVS) dilution tunnel. The engine exhaust was then mixed with dilution air and sampled by the LII and a conventional double-dilution PM sampling system. This PM sampling system consisted of a secondary dilution tunnel that was used to further dilute the previously diluted exhaust sample from the CVS tunnel. The double-diluted PM sample was collected on a pair of 70 mm Teflon-coated glass-fiber filters of the type TX40HI20WW made by Pallflex. The filter pair was arranged such that one of the filters was the main filter and the other a backup situated a few inches behind the first filter. The double-diluted exhaust sample passed through both filters. The majority of the particulate matter was trapped by the first filter, and the backup filter captured most of the remaining PM. The total filtering efficiency is expected to be greater than 95%. Both the primary and secondary dilution air were filtered prior to mixing with the exhaust sample, but no HEPA or hydrocarbon filtration was carried out. The dilution air temperature was maintained between 68 ^oF and 86 ^oF. The filter temperature was controlled to be at or below 125 ^oF during sampling.

The weight of particulate matter collected on the filter pair was determined gravimetrically after the filter sample was conditioned in a temperature and humidity controlled filter weighing room where the blank filter pair had previously been weighed. The volatile material on the filter pair was then extracted using a supercritical fluid extraction technique with CO_2 as the extraction medium. The extracted filter pair is then reweighed to determine the weight of volatile material that was removed. Because the extraction process also removes a measurable amount of material from the filter media that was used to collect the PM, it is necessary to account for this parasitic loss. The contribution cannot be determined from the filter pair used to collect the particulate sample, so an estimate is made by extracting material from filter blanks of the same batch of filters. Because the uniformity of the filter media also varies within a specific batch, uncertainties in the estimate of parasitic losses causes inacccuracies in dry PM measurements determined by this extraction technique. The error that is introduced depends upon the amount of true volatile PM on the filter. If it is comparable to the amount extracted from blanks, there is a potential for large errors in estimating the dry PM amount.

CUMULATIVE RESULTS

Fig. 1 compares cumulative LII measurements (i.e., time-integration of the instantaneous LII signal) with total gravimetric results for 363 steady-state mode tests. Because LII measures only EC, and not the total PM, we do not expect this to correlate well, but present the measurements to illustrate the differences between the total and dry PM correlations as contrasted to the “scatter” in the data. Fig. 2 compares the same LII data with the dry gravimetric results for the 142 tests for which extraction was performed. The most significant difference in the general behavior between the dry and total data is the increased scatter in the dry measurements in the direction of over-estimation of the volatile fraction.

The non-linear relationship in Fig. 2 between LII measurements of the EC volume fraction and the dry gravimetric measurements is somewhat unexpected, since it would seem reasonable to expect the density of dry PM to be relatively constant at approximately

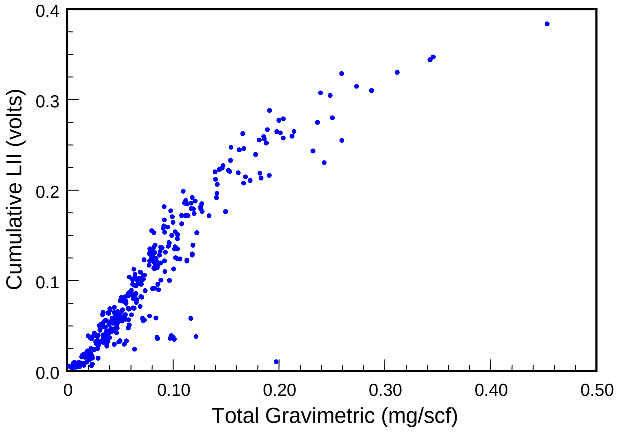

Fig. 1 Correlation between LII and total gravimetric measurements for 363 steady-state mode tests.

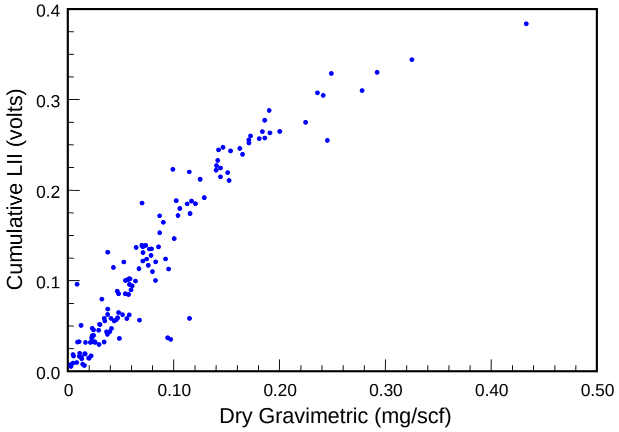

Fig. 2 Correlation between LII and dry gravimetric measurements for 142 steady-state mode tests.

1.9 mg/m^3. Nonlinear response of the photomultiplier tube at high voltage levels has been accounted for in the data analysis.

Snelling et al. [14] conducted a similar study, and report a linear relationship between LII and total gravimetric measurements for a range up to 1.0 mg/scf. Unfortunately, there are too many differences between their experiment and ours to offer an explanation for the difference in observed trends.

There is no inherent reason to expect the gravimetric measurements to be erroneously high at high PM levels. The most plausible cause for the LII to be low at high PM levels is that the exhaust flow is no longer "optically thin". This would attenuate the incident laser excitation and the incandescence, with an expected exponential decrease in LII signal level with increased opacity. Unfortunately, this can be verified only by a return visit of the Sandia experiment to the Cummins test facility.

The next most plausible cause would be the effect of increased volatile organic fraction (VOF) at high PM levels on the LII signal strength. Because the laser energy was optimized for a "typical" test condition, and then kept constant for all tests, the LII signals will be low for deviations in the VOF: For tests with lower VOF, the extra laser energy will go into sublimation of the EC, reducing the volume fraction; for tests with higher VOF, the extra energy needed for desorption will result in lower particle temperatures and, hence, lower LII signals. However, the differences shown in Fig. 3 between total and dry PM do not support this mechanism as the source of the nonlinear behavior seen in Fig. 2.

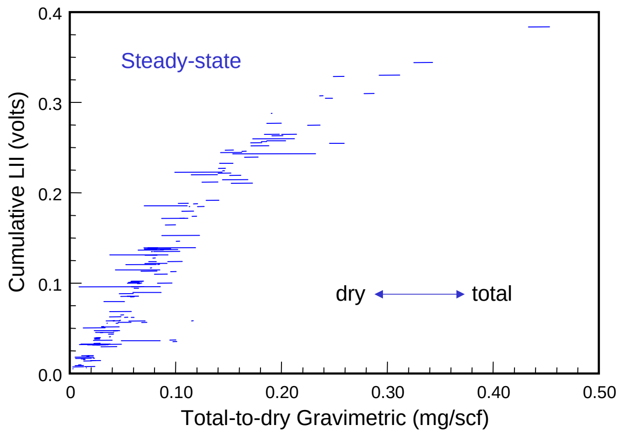

Fig. 3 Shift in the gravimetric mass from total to dry PM.

While it would be desirable to understand the physical processes that caused the nonlinear behavior in Fig. 2, this is not critical for the quantitative application of LII. In Fig. 4 we show an empirical least-squares curve fit to the same data, except we have excluded the obvious statistical outliers (6 points out of 142). The linear portion of the curve fit intercepts the abscissa at 0.0071 mg/scf, which we feel is acceptably close to the origin considering the uncertainties in the dry gravimetric measurements discussed earlier.

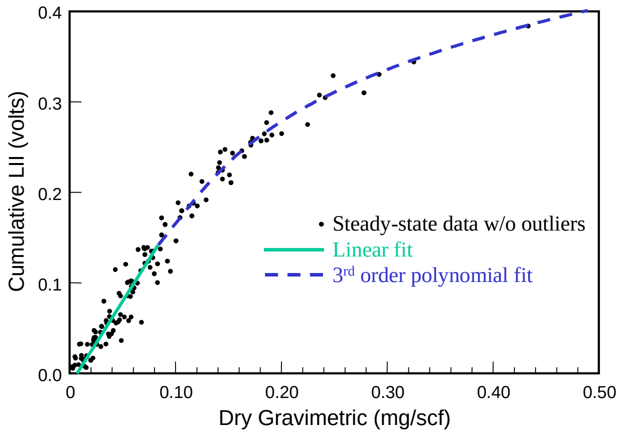

Fig. 4 Calibration of the LII measurements using the dry gravimetric data.

Shown in Fig. 5 are both the steady-state and transient data with the calibration applied to the LII measurements. All of the transient data are shown, for which the full-scale range is limited to 0.12 mg/scf. Recall that these are integrated results, whereas the instantaneous peak values often extend far into the nonlinear region of LII/gravimetric correlation. The solid lines in Fig. 5 are linear least-squares fits to the data (again excluding the 6 outliers in the steady-state measurements). We consider the similarity of the two slopes to be acceptable, considering the uncertainty in the dry gravimetric results at low filter loadings.

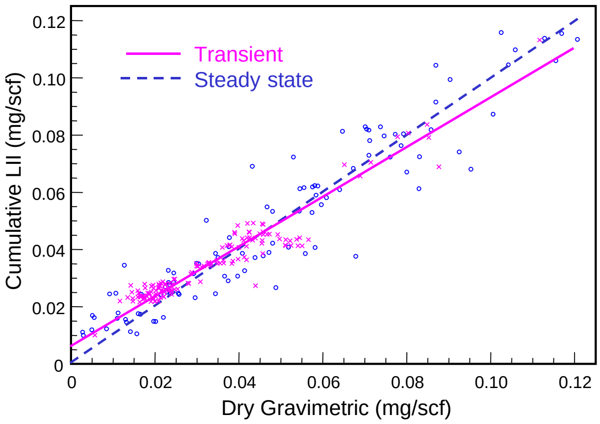

Fig. 5 Correlation between LII and dry gravimetric data for steady-state tests with less than 0.12 mg/scf and all 147 transient tests.

TIME-RESOLVED RESULTS

A typical result for time-resolved LII measurements for the transient FTP is presented in Fig. 6, together with concurrent measurements of the engine speed and load. The PM transients for this heavy-duty transient FTP cycle are far more severe than observed for the light-duty FTP cycle [2], and we believe that the ~3 Hz LII data rate we achieved for these measurements is not really sufficient to capture the peaks and valleys in the LII signal. (As mentioned earlier, our laser runs at 20 Hz, but data acquisition is limited by the download speed of the digital oscilloscope.) However, analysis shows that the lost area from truncation of the peaks has an insignificant effect on the cumulative measurements because of the narrowness of the peaks.

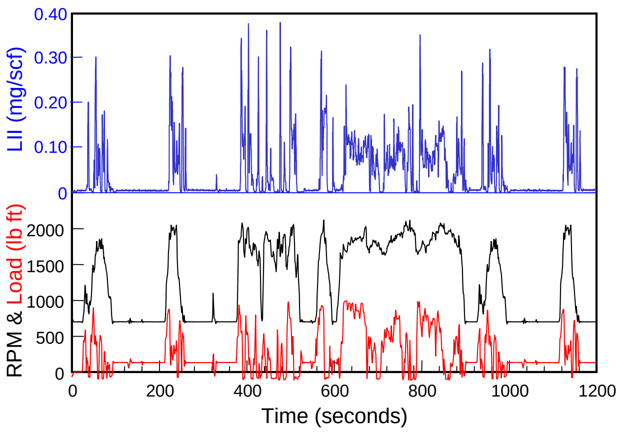

Fig. 6 Typical time-resolved LII measurements for the transient FTP.

In Fig. 7 we show the repeatability of both the transient test itself and the LII measurements for the region

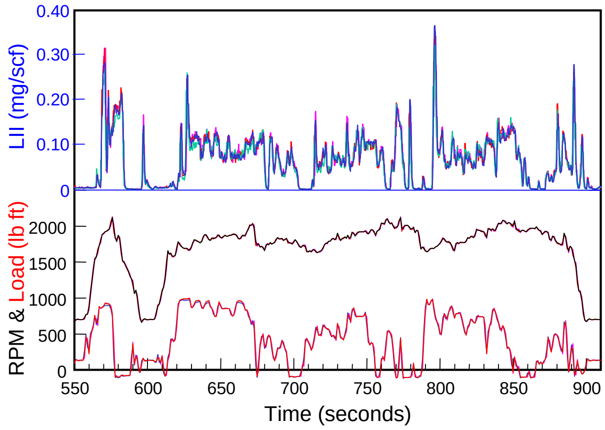

Fig. 7 Comparison of four repeat tests for an expanded region of the transient FTP cycle.

of the test between 550 and 900 seconds. Four repeats are shown, revealing near-perfect repeatability of the engine speed and load, and excellent repeatability for the LII measurements.

The cumulative LII measurements for the four repeat tests are compared with the dry gravimetric results in Fig. 8. The relative rms deviations (rms-deviation/mean) for the LII and gravimetric measurements are 0.026 and 0.074, respectively. It is also interesting to note that there is no correlation in the magnitudes of the two measurements for the repeat tests (i.e., the LII is lowest when the gravimetric is highest, etc.).

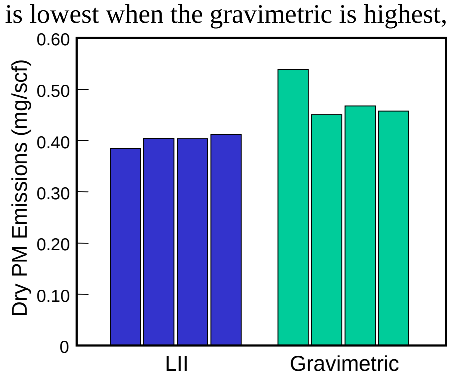

Fig. 8 Comparison of the cumulative dry mass for four repeat tests of the transient FTP.

We have found the time-resolved LII measurements for the steady-state mode tests to be quite revealing about the true "steadiness" of the tests. Three distinct "non-steady" behaviors in PM emissions have been identified: monotonic drift, "bursts", and step changes. Shown in Fig. 9 are examples of monotonic drift for four different test modes. We have chosen to quantify the drift by the expression

$$\text{Percent drift} = \frac{\int_{250}^{300} Mdt - \int_{0}^{50} Mdt}{\int_{0}^{300} \frac{Mdt}{6}} \times 100$$

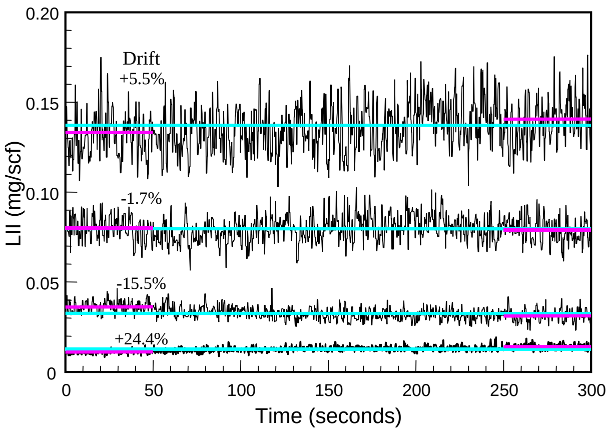

Fig. 9 Examples of time-resolved LII measurements for steady-state mode tests.

where M is the mass concentration. In essence, this is the relative change between the first and last 50 seconds of the test. Because we have normalized by the average value, the percent drift is often exacerbated at low PM levels, as seen in Fig. 9.

In Fig. 10 we show the percent drift for the steady-state mode tests as a function of the cumulative LII measurements. The two data circled correspond to the lower curves in Fig. 9. Plotted in this manner, the increase in percent drift with decreasing PM level is clearly evident.

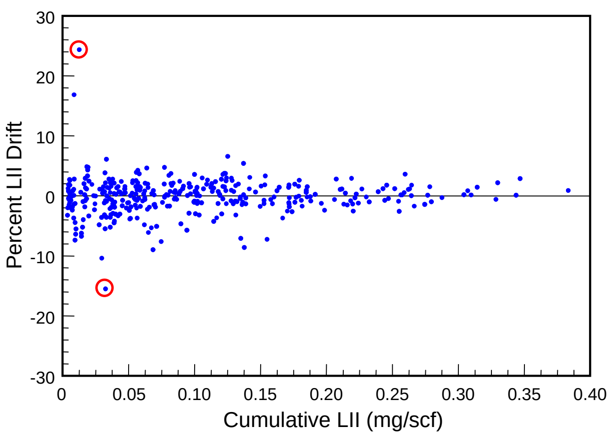

Fig. 10 Percent drift in the time-resolved LII measurements during steady-state mode tests.

The second form of unsteadiness we observed in the steady-state mode tests was sudden, very short-duration bursts of high PM levels, as shown in Fig. 11. We also show corresponding opacimeter measurements made concurrently on the raw exhaust, to confirm that the burst in the LII measurements is not due to a single, large particle that just happened to find its way into the LII probe volume. We believe that these bursts can only be explained by a momentary increase in PM concentration, such as would be caused by a single-injection abnormality. While the percent drift in this test is +3.8, the mass in the burst is too small to make a significant contribution. We observed several occurrences of these

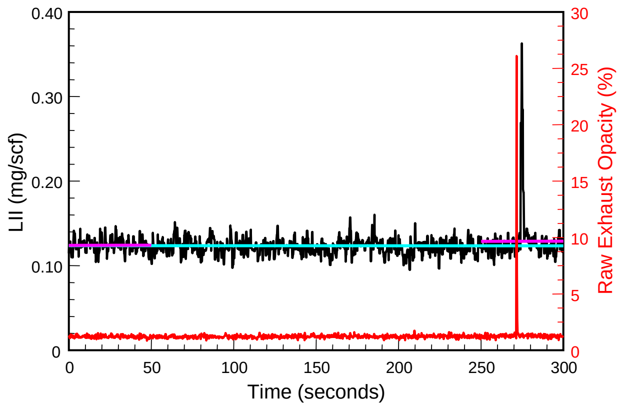

Fig. 11 Example of a PM “burst” detected by both LII and an opacimeter.

bursts, but did not systematically search all 363 tests to determine the actual frequency.

The third type of unsteadiness occurred in the form of abrupt step changes in the PM concentration, as shown in Fig. 12. We again show opacimeter measurements to confirm similar behavior between raw engine out and sampling from the CVS. It appears to us that this is a bimodal phenomena, in that several times the PM level returns to the lower value for short periods (at 105, 155, 180, and 300 seconds) after the initial step change to the higher level at 60 seconds. The percent drift for this test is +32.4, which is off the scale of Fig. 10. Like the bursts, we observed several tests with step changes, but did not search for their actual frequency of occurrence. However, unlike the drift and burst abnormalities, where the impact on the gravimetric measurements will usually be small, a step change as shown in Fig. 12 should be considered an invalid test and excluded from further analysis.

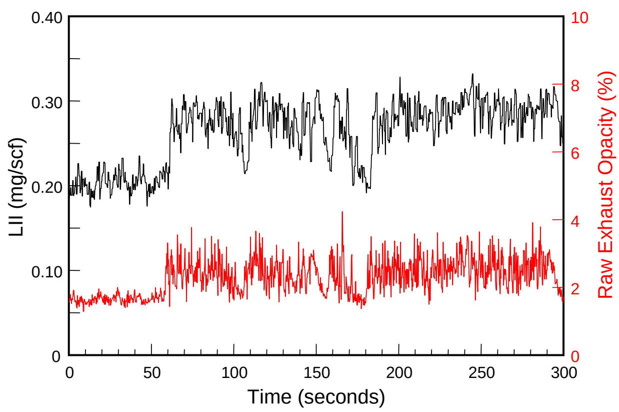

Fig. 12 Example of a step change in PM concentration for a steady-state mode test.

CONCLUSIONS

We have shown that a unique and powerful advantage of LII over most other PM measurement techniques is the capability to run unattended for extended periods of time. An active air-purge system is required to keep the optical-cell windows clean, but this is easily implemented. The remaining factor limiting uninterrupted operation is the laser flashlamp lifetime, which is on the order of at least several months. It would also be trivial to extend the effective lifetime by having the central data acquisition system enable the flashlamp only during actual test periods.

While the demonstration presented herein was for diluted exhaust extracted from a CVS tunnel, this LII system will work equally well on raw exhaust. In fact, we believe that the most useful application of LII will be in engine development, enabling the real-time monitoring of PM emissions for optimization of in-cylinder combustion and aftertreatment performance. It is common for engines to be mapped unattended using gaseous emissions. Smoke opacity has been the normal gage of PM emissions until now. However, PM emissions levels for the 2007 and beyond are at least an order of magnitude below today's levels, and current opacity meters will not be effective at those levels. LII will allow the unattended mapping of PM emissions from these heavy duty diesel engines, making it possible for an engine development engineer to get a full complement of gaseous and dry PM emissions for analysis.

The main disadvantage of LII is that it measures the EC volume fraction, and not the regulated metric of total PM mass. However, we have shown that steady-state mode tests can be used to calibrate LII using dry gravimetric measurements, and then be successfully applied to the transient cycle tests.

Time resolved LII measurements of transient emissions tests would be a very useful tool in engine development. More and more engineers are requesting transient emissions capability in previously steady-state test cells. Engine test cell control and data acquisition systems have responded to provide this capability. LII would be a useful tool to monitor transient emissions PM characteristics.

Finally, time-resolved LII measurements of steady-state mode tests reveal three forms of unsteadiness that we have labeled as drift, bursts, and step changes. In some cases these can be severe, suggesting that LII could be useful for identifying gravimetric test results that could be misleading and should be rejected.

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